



Final Technical Report

Covering the Period March 1972 through April 1977

April 1977

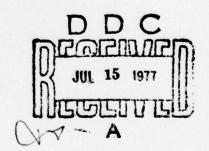
# INVESTIGATION OF A PHOTODICHROIC MATERIAL FOR HOLOGRAPHIC STORAGE AND RECOVERY

By: MATT LEHMANN

Prepared for:

DEPARTMENT OF THE NAVY
OFFICE OF NAVAL RESEARCH
INFORMATION SYSTEMS BRANCH
CODE 430
ARLINGTON, VIRGINIA 22217
Attention: Joel Trimble

CONTRACT N00014-72-C-0260





STANFORD RESEARCH INSTITUTE Menlo Park, California 94025 · U.S.A.

DISTRIBUTION STATEMENT A

Approved for public telease,
Distribution Unlimited

MAY 1 8 1977

.... . ....

Unclassified
SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM	
1. REPORT NUMBER	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATA	
4. TITLE (and Subtitle)	(9)	5. TYPE OF REPORT & PERIOD COVERED	
INVESTIGATION OF A PHOTODICHROIC MA	TERIAL FOR	Final Technical Report,	
HOLOGRAPHIC STORAGE AND RECOVERY.		March 1972 the	agh April 197
		6. PERFORMING ORG.	REPORT NUMBER
		SRI-PYU-1777	
7. AUTHOR(s)		8 CONTRACT OR GR	ANT NUMBER(s)
Matt/Lehmann		Contract N00014-72-C-0260	
		Contract Nood!	1-72-0-0260
9. PERFORMING ORGANIZATION NAME AND ADDRE	ESS	10. PROGRAM ELEME	NT, PROJECT, TASK
Stanford Research Institute V		AREA & WORK OF	WIT NOWINERS
333 Ravenswood Avenue			
Menlo Park, California 94025 11. CONTROLLING OFFICE NAME AND ADDRESS		12. REPORT DATE	13. NO. OF PAGE
office of Naval Research	(1)	Apri 1977	39
Information Systems Branch		15. SECURITY CLASS.	(of this report)
Code 430, Arlington, Virginia 22217		Unclassified	TX 29 /
14. MONITORING AGENCY NAME & ADDRESS (if dif		(	WOIP.
		15a. DECLASSIFICATION	ON / DOWNGRADING
		SCHEDULE	
	1 for	STATEMENT A  public release;	
	Distribution	n Unlimited	
17. DISTRIBUTION STATEMENT (of the abstract entere	ed in Black 20, if different f	rom report)	
18. SUPPLEMENTARY NOTES			
19. KEY WORDS (Continue on reverse side if necessary	and identify by block number	er)	
Optical storage			
Alkali halides			
Photodichroic crystals			
Holographic recording			
20. ABSTRACT (Continue on reverse side if necessary an	id identify by block number)		
Sodium fluoride (NaF), a space			stal when
irradiated by high-energy electrons			
tion bands that can be modified by			
uv light at room temperature. The			
very high resolution polarizable ab			
			-
and is therefore capable of storing The material is capable of write, e	and recovering in	formation hologr	aphically.

DD 1 FORM 1473 EDITION OF 1 NOV 65 IS OBSOLETE

Unclassified
SECURITY CLASSIFICATION OF THIS PAGE (When Data Enter )



SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

19. KEY WORDS (Continued)

20 ABSTRACT (Continued)

We have identified a refractive-index shift at the base of the absorption zones, which suggests that phase holography is possible in this material, with attendant improved diffraction efficiencies.

Modification of color-center formation by selected ion implantation indicates that higher sensitivity and diffraction efficiencies are possible without sacrificing the desirable properties of the basic material. Fourier-transform holograms as well as multiple gratings have been recorded and recovered. This material has satisfactorily demonstrated that it is a prime candidate for holographic storage where real-time write, read, and erase are specified requirements.

DD 1 FORM 1473 (BACK) .
EDITION OF 1 NOV 65 IS OBSOLETE

# CONTENTS

LIST	OF I	LLUSTRATIONS	
I	INT	RODUCTION AND SUMMARY	:
II	EXP	ERIMENTAL PROGRAM	5
	Α.	Information Storage in Photodichroic Materials	5
	в.	Holographic Storage and Recovery	8
		1. Bragg Suppression	11
		2. Refractive Index Modulation as a Function of M Center Absorption	12
	c.	Ion Implantation in Sodium Fluoride	16
	D.	Technique for Image Enhancement and Improved Writing Sensitivity in Alkali Halide Crystals	17
	E.	Crystal Preparation	24
	F.	Effects of Controlled Temperature and Electric Fields on Sensitivity	26
III	CON	CLUSIONS	35
IV	REF	ERENCES	37



## ILLUSTRATIONS

1.	Harshaw-grown NaF Implanted with Lithium Ions	6
2.	Refractive Index as a Function of M-Center Absorption	7
3.	Formation of M Centers in an Alkali Halide Crystal	9
4.	Holographic Setup for Write/Read on Alkali Halide Crystal	10
5.	Diffraction Efficiency as a Function of Read Wavelength	13
6.	Elliptically Polarized Light by Aligned M-Center Absorption	15
7.	Setup for Measurement of Refractive Index Shift as a Function of M-Center Absorption	15
8.	Optical Density as a Function of Dosage for Selected Ions	18
9.	Li Ion-Implanted NaF Absorption Spectra after Exposure to Polarized uv.	19
10.	1 cm Square NaF Crystal with 400 Independent Sites	20
11.	Photodichroic Alkali Halide as an Incoherent/Coherent Processor	22
12.	Effect of M-Center Absorption on Polarization in NaF Crystal	23
13.	High-Contrast Image Produced by Enhancement Technique	25
14.	Effect of Temperature Rise on M-Center Absorption Peak	27
15.	Shift in M Center Peak	28
16.	Specimen Holder for Temperature Control	30
17.	Effect of Temperature and Radiation on M Center Peak	32
18.	Specimen Holder for Electric Field Experiment	33

### I INTRODUCTION AND SUMMARY

Investigations of photochromic information storage in certain selected alkali halides led to the observation that a distinct dipole moment was associated with on-center absorption. This observation implies that orientation of the M centers and the absorption spectra depends on the polarization of both the write and the read illumination. Such a material is more appropriately termed photodichroic. These preliminary studies, conducted by the Naval Research Laboratories, suggested the application of photodichroic materials to the field of holographic information storage by virtue of their very high resolution capability.

The demand for both very large memories and rapid access time favors the use of high density optical memories. The ideal memory must have write, read, and erase modes; a nondestructive readout with a permanent memory when required; very high resolution and packing densities; good sensitivity for rapid write capability; and accessibility to a specified bit without prior knowledge of its precise location in the memory. In addition, it is desirable, though not essential, that the memory function in all modes at ambient room temperature. It would also be advantageous for the memory to be relatively immune to damage by electrical surges or nuclear radiation; in fact, this requirement may be a priority for certain military applications.

Studies initiated in 1972 and continuing to the present have demonstrated that the photodichroic alkali halide sodium fluoride (NaF) exhibits all the desirable traits for holographic or image storage with the exception of good sensitivity. It has been established that NaF can be used for information storage either as an image that

can be read with suitably polarized light at the wavelength  $(\lambda_r)$ , or from holographic storage using a polarized reference centered at the peak of the M band absorption spectrum or at the edge of the M band for refractive readout (dielectric hologram). Irradiation of the NaF crystal with penetration radiation (high-energy x rays, gamma rays, or electrons) produces color centers throughout the depth of the material, whereas ion-implantation techniques produce a thin layer of color centers on the surface of the material due to the relatively short range of the ions. The color centers formed in NaF by irradiation with ionizing radiation or high-energy particles can have the absorption and refraction modified and spatially modulated by illumination with polarized light at the writing wavelength ( $\lambda_r$ ) and detected at a nondestructive read wavelength ( $\lambda_r$ ).

Grinding and polishing are essential to ensure satisfactory image quality particularly for the ion-implanted specimens, because the color center depth is very shallow (2 to 5  $\mu$ m). Furthermore, since these materials were being studied for device applications, it was essential to determine the temperature range within which the color centers would maintain their integrity. This led to the discovery that writing at slightly elevated temperatures would increase the sensitivity by causing the storage sites to be more optically active.

This report describes an experimental program to evaluate holographic diffraction efficiency, effect of impurities on modulation fatigue, influence of surface defects, optimum coloration for both ion-implanted and bulk irradiated specimens, modifications for enhanced sensitivity, and other parameters concerned with the application of photodichroics in optical memories and optical computers.

#### II EXPERIMENTAL PROGRAM

## A. Information Storage in Photodichroic Materials

The atomic structure of typical ionic compounds, such as alkali halides, is that of a face-centered cubic lattice. In the pure state, these substances are transparent over a wide wavelength spectrum. For sodium fluoride the pure crystals transmit light from 130 nm to 12  $\mu$ m. The color-center phenomena may be explained by the existence of impurities or structural imperfections in the crystal lattice, which create electron hole traps. Charge carrier trapping resulting from irradiation with ionizing radiation or high-energy particles causes absorption bands to form in these otherwise transparent crystals.

What makes these materials ideal candidates for high resolution information storage is their unique ability to vary wavelength of the light absorption peak as a function of the amplitude of the polarization vector of the incident illumination. When polarized light of the correct wavelength ( $\lambda_{\rm w}$ ) is used to write in the prepared crystal, absorption increases at the read wavelength ( $\lambda_{\rm r}$ ) and decreases along the orthogonal axis as shown in Figures 1 and 2. Spatial modulation of the absorption, therefore, makes it possible to store information (and attendant refractive index shift) in the crystal.

Sodium fluoride (NaF) was selected for this effort because F ( $\lambda_w$ ) and M ( $\lambda_r$ ) colors centers are readily formed by irradiating the crystals with high-energy particles, x rays, or gamma rays at room temperature. Additional M centers can be formed from existing F centers with polarized uv light at a wavelength of approximately 350 nm and stored also at room temperature. The M and F centers in NaF are particularly attractive for use in an operational device as they are formed at room temperature.

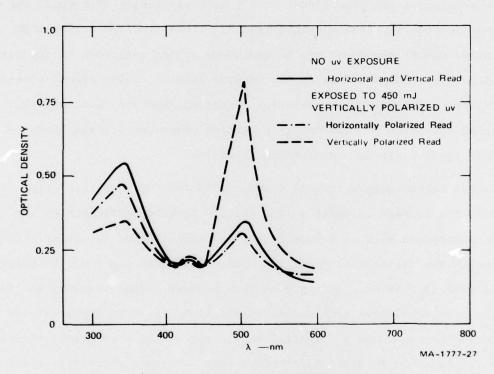


FIGURE 1 HARSHAW-GROWN NaF IMPLANTED WITH LITHIUM IONS 1.85 MeV -  $10^{14}/\text{cm}^2$ 

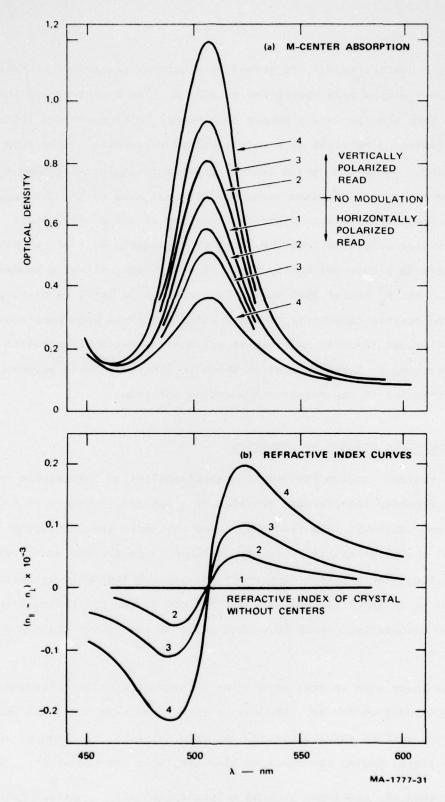


FIGURE 2 REFRACTIVE INDEX AS A FUNCTION OF M-CENTER ABSORPTION

The F centers in NaF are formed by electrons trapped at a negative ion vacancy with a peak absorption at 343 nm. The M centers and the F centers form simultaneously during high energy bombardment and later, when illuminated by light of a wavelength corresponding to the peak of F-center absorption. These absorption centers constitute nearest-neighbor F centers (trapped electrons) oriented along the diagonal axes of the face-centered cubic crystal (Figure 3). These M-center peaks are at 505 nm and thus substantially separated from the F-center absorption so that the read wavelength ( $\lambda_{\Gamma}$ ) does not modify the F or  $M_{\Lambda}^{\dagger}$  centers, allowing nondestructive readout. The  $M_{\Lambda}^{\dagger}$  center lies under the F-absorption band, permitting the erase and rewrite capability, because after F centers have been converted to M centers it is still possible to write with F-band illumination ( $\lambda_{W}$ ) resulting in large changes in M-center absorption with correspondingly small variation in the F-center absorption spectrum.

## B. Holographic Storage and Recovery

To evaluate sodium fluoride for applicability to holographic recording, we have recorded interference gratings at a spatial frequency of 1000 cycles/mm. Although this frequency in no way tests the resolution limit of the material, it is in the probable range for any practical application. The gratings were recorded with a holographic apparatus using a coated prism to divide the circular pattern of the collimated beam into two semicircles which were then superimposed on the crystal surface (Figure 4).

The laser used in this effort was a Coherent Radiation Laboratories Model 52G modified for uv. The two uv lines used were 351.1 and 363.8 nm which were emitted simultaneously. An external prism was used to separate the two lines, making the 351.1-nm line available for exposing the NaF.

To read the gratings, we used a Spectra Physics, Model 262 argon

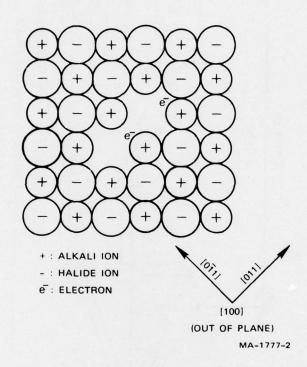
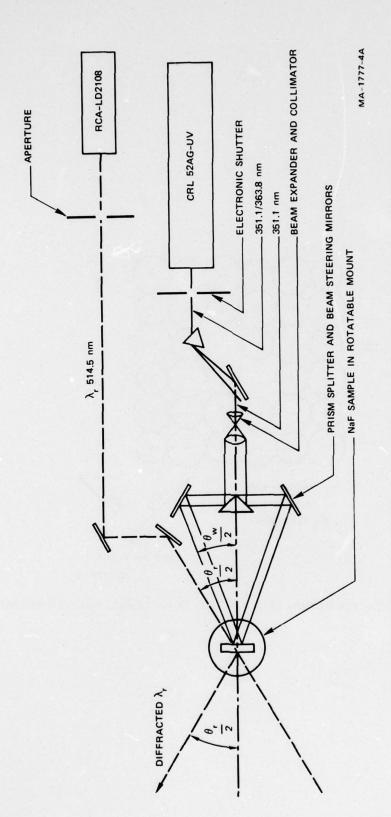


FIGURE 3 FORMATION OF M-CENTERS IN AN ALKALI HALIDE CRYSTAL



1 .

SCHEMATIC OF HOLOGRAPHIC APPARATUS FOR WRITE/READ ON ALKALI HALIDE CRYSTAL FIGURE 4

laser that provided approximately 5 mW at 514.5 nm and 10 mW at 488.0 nm. The geometry was arranged to permit writing with 351.1 nm followed by immediate reading with 514.5 nm without disturbing either optical system. It was found necessary to include an aperture on the read laser to prevent the uv in the plasma glow from erasing the gratings. Both lasers were polarized vertically.

## 1. Bragg Suppression

Thick holographic gratings are characterized by Bragg-angle sensitivity for efficient diffraction. For holographic recordings, the readout illumination is usually incident at the identical angle and of the wavelength as the reference illumination during recording. Deviation from the recording angle will reduce the coherent reconstruction of the image, which for a simple grating results in a reduction in the diffraction efficiency. This study depended on the  $\mathbf{M}_F^+$  absorption band  $(\lambda_W)$  for writing, and the M absorption band  $(\lambda_W)$  for reading. Since the reference wavelength for readout was not the same as  $\lambda_W$ , it was necessary to modify the read angle to correct for the wavelength difference. The effects of wavelength changes on grating responses are similar to the effects of angular changes, so that changes in angle can be used to compensate for wavelength changes. This interdependence between angle and wavelength is expressed by the equation:

$$\frac{\theta_{r}}{2} = \sin^{-1}\left(\frac{\Lambda}{2} \lambda_{r}\right) \tag{1}$$

where

 $\Lambda$  = Spatial frequency of the grating established by the angle and wavelength during recording

 $\theta_r$  = Read angle (degrees)

 $\lambda_n = \text{Read wavelength (nm)}.$ 

Although this relationship is appropriate for all holographic recording, the precision to which this angle must be controlled depends on the spatial frequency of the grating and the grating thickness

$$\Delta \boldsymbol{\theta} = \Lambda/t \tag{2}$$

where

 $\Delta \theta$  = Angle change that will completely suppress reconstruction--mrads

t = Grating thickness in micrometers when the spatial frequency  $\Lambda \; \text{is in cycles/mm.}$ 

The specimens used in this study were approximately 0.50 mm thick and therefore highly dependent on angular orientation. The measured angle for complete Bragg suppression of the diffracted image was 3.4 mrad, which, by Equation (2), predicts a grating thickness of 0.294 mm, somewhat less than the actual thickness. This discrepancy can be explained by intensity reduction as a result of absorption, or by beam decoupling caused by the effect of phase-shifting on interference as a function of the depth of writing. Four holograms were recorded sequentially at 10-mrad separation. The first hologram in this series was partially erased as the fourth was being recorded. There was no apparent interference between adjacent holograms during recovery.

# 

Early in this study of holographic storage we discovered that reconstruction with a wavelength at the edge of the M center absorption spectrum greatly improved the diffraction efficiency (Figure 5). This discovery verified that the refractive index shift is large enough to expect the improved diffraction efficiency attributed to mixed holographic gratings [Equation (3)].

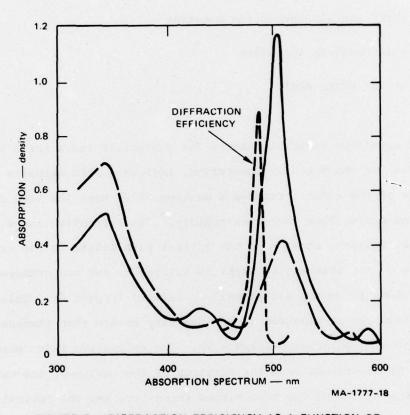


FIGURE 5 DIFFRACTION EFFICIENCY AS A FUNCTION OF READ WAVELENGTH

$$\eta = \left[ \sin^2 \left( \pi \eta_1 d / \lambda \cos \theta_0 \right) + \sin h^2 (\alpha_1 / 2 \cos \theta_0) \right] \exp(-20 d / \cos \theta_0). \quad (3)$$

- η is the diffraction efficiency
- $\eta_1$  and  $\alpha_1$  are the amplitudes of the modulation of the refractive index and absorption constant
- a is the average absorption constant
- d is the grating thickness
- 6 is the Bragg angle

The anomalous dispersion curve for refractive index shift vanishes at the peak of the M-center absorption, increases with opposite signs on each side of the center, reaches a maximum value near the base of the absorption curve, then decreases rapidly. As refractive index changes are, by definition, changes in the optical path length in the crystal, the phase of the transmitted light is shifted by the two orthogonal polarizations to emerge elliptically polarized (Figure 6). This polarization can be measured as an intensity change when examined through crossed polarizers (Figure 7). The refractive index changes can then be determined from the rotation of the analyzer, the maximum and minimum values of the transmitted intensity, and the optical density of the M-center absorption for the two orthogonal polarization axes of the crystal by:

$$n=n_{\perp} = \frac{\lambda}{2\pi d} \cos^{-1} \left\{ \left( \frac{\frac{a}{a} + \tan \theta}{\frac{a}{1}} \right)^{2} - \left[ \tan^{2} \theta + \left( \frac{\frac{a}{a}}{a} \right)^{2} \right] \frac{a_{\perp}^{2}}{a_{M}^{2}} \right\}$$

$$\frac{2\left(\frac{a}{a}\right)}{a} \left( \frac{1 + \frac{a^{2}}{a}}{a^{2}M} \right) \tan \theta$$

$$(4)$$

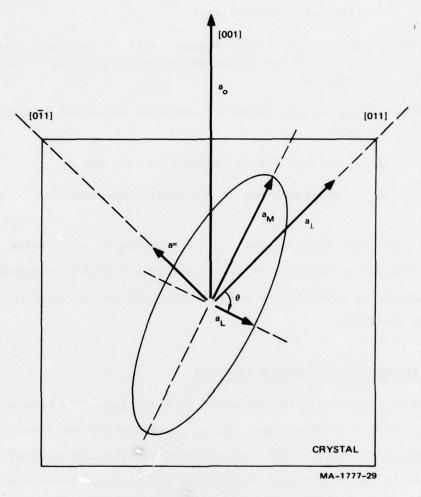


FIGURE 6 ELLIPTICALLY POLARIZED LIGHT BY ALIGNED M-CENTER ABSORPTION

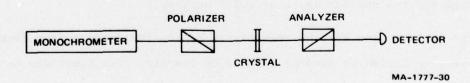


FIGURE 7 SETUP FOR MEASUREMENT OF REFRACTIVE INDEX SHIFT AS A FUNCTION OF M CENTER ABSORPTION

- $\frac{n}{n} = \frac{\text{and } n}{n}$  are the refractive indices for the horizontal and vertical polarized axes
- a and a are the amplitudes measured with the analyzer oriented along the two orthogonal polarization axes of the crystals
- $a_L$  and  $a_M$  are the amplitudes of the major and minor elliptical axes
  - λ is the wavelength of light in free space
  - d is the thickness of the crystal specimen.

The refractive index change  $(n_{=}-n_{1})$  in the crystal varies linearly with the modulation in M-center absorption as a result of irradiation with polarized uv light in the F-absorption band and is shown in Figure 2 for a NaF specimen.

## C. Ion Implantation in Sodium Fluoride

Thin holograms have the advantage of being less sensitive to the angular precision of the reconstructing reference and are more efficient in the absorption mode. The concentration of radiation colored centers in the alkali halide crystals varies with the depth of penetration and is a function of accelerator energy and exposure time. The absorption spectra for the ion-implanted crystals are identical with those in the bulk-colored specimens except that the absorption is less, as would be expected for the shallow depth of color centers.

To evaluate the writing sensitivity of ion-implanted sodium fluoride crystals, we implanted several ions using the Air Force Cambridge Laboratories ion-implantation facility. All the heavier ions (i.e., boron carbon, fluorine, and oxygen) were implanted at 3 MeV to a fluence of from 5 x  $10^{13}$  ion/cm<sup>2</sup> to  $10^{15}$  ion/cm<sup>2</sup>. The lithium ion specimen was implanted

at 1 MeV to a fluence of  $5 \times 10^{14}$  ion/cm<sup>2</sup>. All comparative curves shown are for specimens having a fluence of  $5 \times 10^{14}$ /cm<sup>2</sup> to conform with the lithium ion-implanted specimens. Lower ion dosage with the heavy ions produced fewer color centers so the absorption spectra were even less informative (Figure 8). The characteristic F and M center absorption spectra were observed for all specimens. It is fairly evident from the data of Figure 8 that the lighter ions produced a higher concentration of both F and M centers. With the exception of the lithium implanted crystals, attempts to write in the ion-implanted specimens resulted in depletion of F center absorption without any increase in M band absorption. Attempts to erase and rewrite were not successful. It is probable that no  $M_A^+$  centers were being formed, which would account for both the nonreversible reduction of the F band absorption and the inability to erase and rewrite with polarized uv.

As a result of this analysis, we continued our efforts using Li ion-implantation at 1.9 MeV to a dose of  $10^{14}$  to  $10^{15}$  ions/cm<sup>2</sup>, resulting in satisfactory F-band absorption peaks at 340 nm ( $\lambda_{\rm W}$ ) and a smaller M-center peak at 505 nm ( $\lambda_{\rm F}$ ). The depth of color centers in polished Li ion-implanted specimens has been determined to be 3 to 4  $\mu$ m. After exposure to polarized uv, the M center absorption increases as the F absorption decreases. The change in absorption spectra after exposure to uv is shown for comparison (Figure 9). We have demonstrated that ion-implantation can be used to produce spatially isolated storage sites that would make it possible to store more than 400 independent images in a centimeter square crystal (Figure 10).

# D. Technique for Image Enhancement and Improved Writing Sensitivity in Alkali Halide Crystals

Analysis of the refractive index shift as a function of color-center absorption in NaF led to the development of a technique that improves

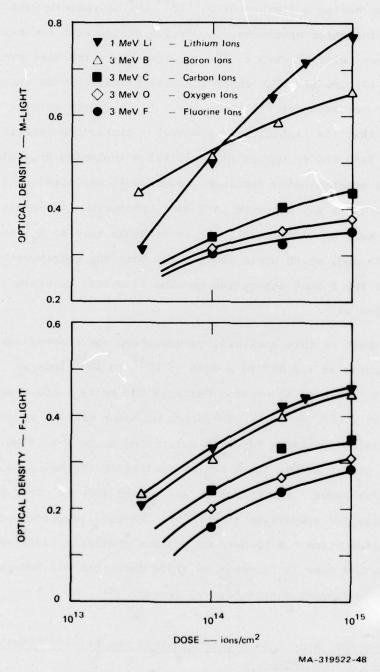


FIGURE 8 OPTICAL DENSITY AS A FUNCTION
OF M-CENTER ABSORPTION FOR
SELECTED LONG IMPLANTATION

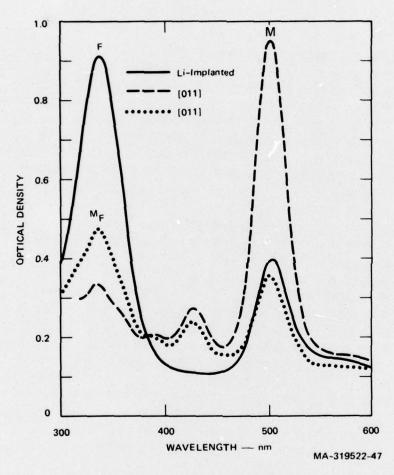


FIGURE 9 CHANGE IN LI ION IMPLANTED NaF ABSORPTION SPECTRA AFTER EXPOSURE TO POLARIZED UV

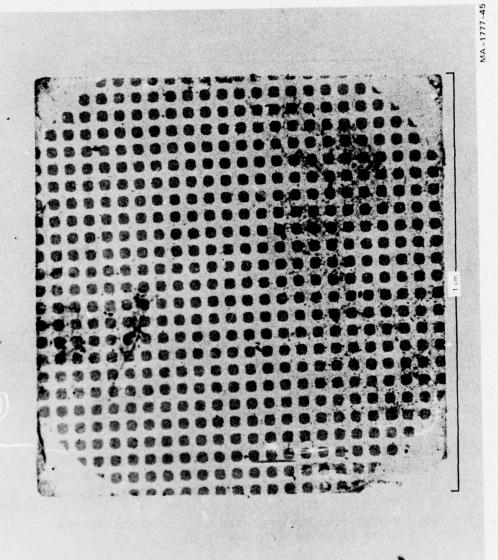
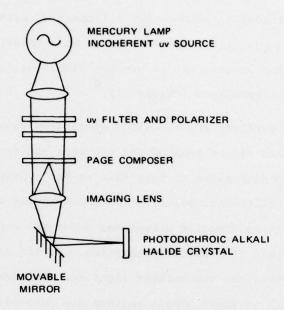


FIGURE 10 ION IMPLANTED CRYSTAL WITH MORE THAN 400 INDEPENDENT STORAGE SITES

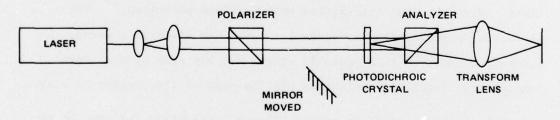
the effective writing sensitivity by an order of magnitude. The formation of high-contrast images in NaF requires approximately 450 mJ/cm<sup>2</sup> of polarized uv light; therefore, any reduction in this energy requirement would constitute an improvement in writing rate. High-contrast images can be achieved without total M-center alignment by using the dichroism of the material to read the image through crossed polarizers. This technique also makes it possible to use the alkali halide crystal as an incoherent/coherent processor (Figure 11).

As mentioned earlier, the anomalous dispersion curve for refractive index shift vanishes at the peak of the M-center absorption band and increases with opposite signs on each side of the center. This would indicate that the elliptical polarization accompanying the refractive index shift rotates in opposite directions about the center and changes direction at the peak of M-center absorption. As the minor axis of the ellipse becomes zero, the transmitted light becomes plane polarized. Also, the M centers in these alkali halides are photodichroic and selectively absorb one of the two rectangular components of ordinary light; therefore, the transmitted beam is plane polarized. The elliptical polarization caused by the anomalous dispersion across the absorption spectrum is a separate effect but can also be used for readout with light of wavelengths off the peak of the absorption curve.

When a light source is passed through a polarizer followed by an analyzer at right angles to the polarizer, no light is transmitted through the analyzer. If we place a specimen crystal in which the M-center absorption is equal for both rectangular polarization axes between the polarizer and analyzer, then the polarization remains unchanged and no light is transmitted by the analyzer. If we irradiate the crystal with polarized uv light so that the M-center absorption is not the same for both axes (i.e.,  $OD_{=} - OD_{=} ^{\ddagger} O$ ), then the polarization of the incident illumination is modified and is transmitted by the analyzer (Figure 12).



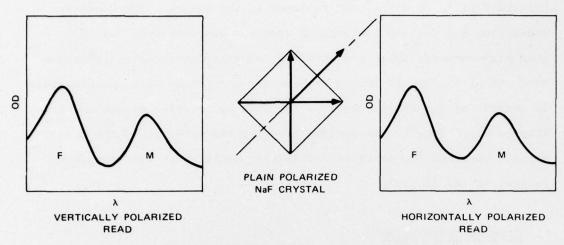
## (a) WRITE MODE INCOHERENT IMAGING



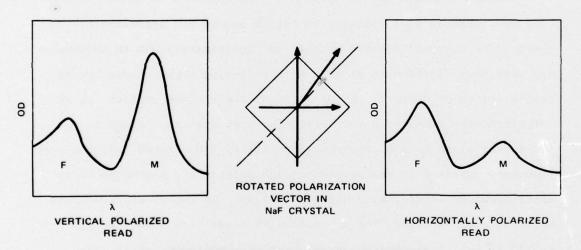
(b) READ MODE COHERENT OPERATION

MA-1777-32

FIGURE 11 PHOTODICHROIC ALKALI HALIDE AS AN INCOHERENT/COHERENT PROCESSOR



(a) M CENTER ABSORPTION UNIFORM FOR BOTH CRYSTAL AXES



(b) AFTER IRRADIATION WITH VERTICALLY POLARIZED uv M Center Absorption is Modified.

MA-1777-33

FIGURE 12 EFFECT OF M-CENTER ABSORPTION ON POLARIZATION IN NaF CRYSTAL

A NaF specimen was irradiated with unpolarized uv and placed between the polarizer and analyzer oriented for minimum transmission. Subsequently, the specimen was irradiated with spatially modulated polarized uv to 50 mJ/cm<sup>2</sup> and replaced in the system. The M-center modulation for the two rectangular axes was approximately 0.15 OD. A good high-contrast image was obtained, demonstrating that this method produces high-contrast images by M-center modulation with approximately 10 percent of the energy required for full orientation of the M-centers (Figure 13). Erasure and cycling are also improved, since the low writing energies for satisfactory imaging minimize optical damage and fatigue in the crystal.

## E. Crystal Preparation

Highest possible purity Harshaw NaF crystals, 1 cm square and up to 2 cm long were exposed to a radioactive cobalt source for approximately 24 hours. The crystals cleaved readily to approximately 300 \$\mu\$m thickness and were then bleached in an oven with a flowing argon atmosphere to remove all color centers. Examination of the cleaved surfaces in an interferometer revealed a very irregular wavy surface. Since it was not essential to have both surfaces parallel, the cleaved crystals were then hand polished to remove all surface defects and inspected in an interferometer to determine surface quality. To remove gross defects we first rough-polished the crystals with Linde Fine Abrasive Type A-5175 in methyl alcohol on AB selected silk polishing cloth, then after the interferometer check, finished the surface by dry polishing the cleaved crystals on microcloth. 11

The prepared crystals were then carefully mounted with a thermal paste in groups of nine in a stainless steel jig designed to fit the mounting facilities of the accelerator target. The crystals in their holder were carefully wrapped and delivered for Li ion-implantation. This

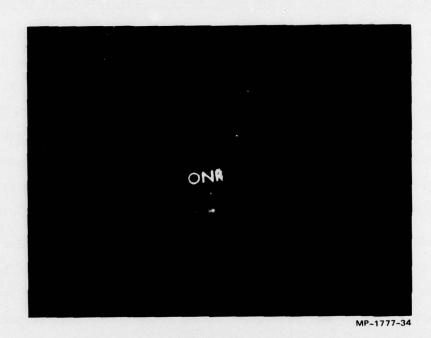


FIGURE 13 HIGH-CONTRAST IMAGE PRODUCED BY ENHANCEMENT TECHNIQUE

procedure assured that the crystals were not touched or handled after polishing.

# F. Effects of Controlled Temperature and Electric Fields on Sensitivity

Experiments with KCl (potassium chloride) indicate that writing sensitivity can be enhanced by elevating the crystal temperature during exposure to the writing cycle and then immediately cooling the crystal to retain the information. With KCl, this experiment required the use of a thermoelectric cooler to maintain a cryogenic storage temperature after writing at room temperature. This approach was easily extended to NaF by using a small electric heater to raise the crystal temperature and cooling it immediately after writing for room temperature storage.

To determine the effect of elevated temperatures on the color centers in a NaF crystal, we constructed a small oven that could be placed in the sample chamber of a Beckman DK2 spectrophotometer to permit monitoring of the absorption spectrum as the crystal temperature was raised (Figure 14). The oven consisted of an outer Teflon casing with quartz windows fitted with gaskets at each end of the Teflon tube. The crystal specimen was supported on a copper disc secured to a pyrophyllite cylinder wrapped with 14 turns of 300  $\mu$ m (12 mil) nichrome wire. A 15V, 0.8A heater element gradually raised the crystal specimen temperature. An iron/constantan thermocouple monitored the crystal temperature, which was recorded on a Leed and Northrop series 620 strip recorder. The M center absorption showed a distinct increase as the specimen was heated to 48°C, followed by a rapid decrease to the preanneal level at 76°C. As the temperature increased, the M center absorption continued to decrease until no color centers remained above a temperature of 205°C (Figure 15) and caused the M center peak to shift from 505 nm to 515 nm. These experiments imply that it may be possible to increase the photosensitivity of the NaF

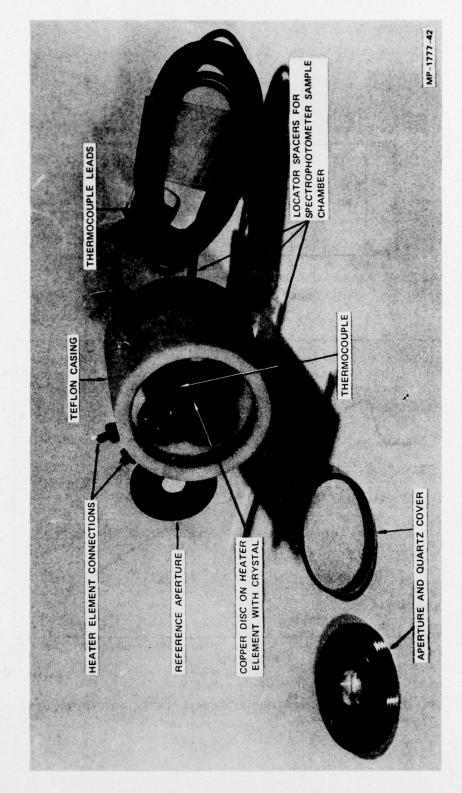
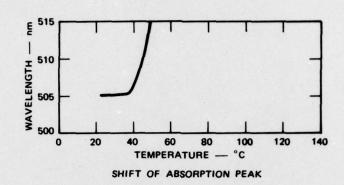


FIGURE 14 OVEN USED TO HEAT SPECIMEN DURING SPECTROPHOTOMETRY



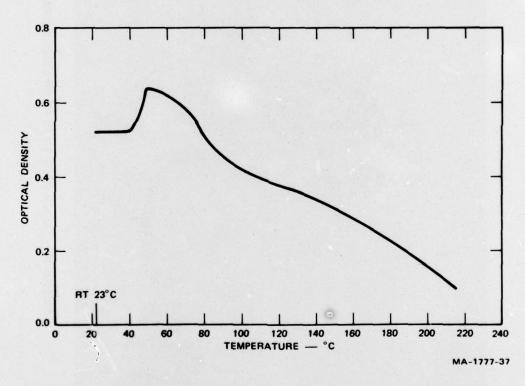


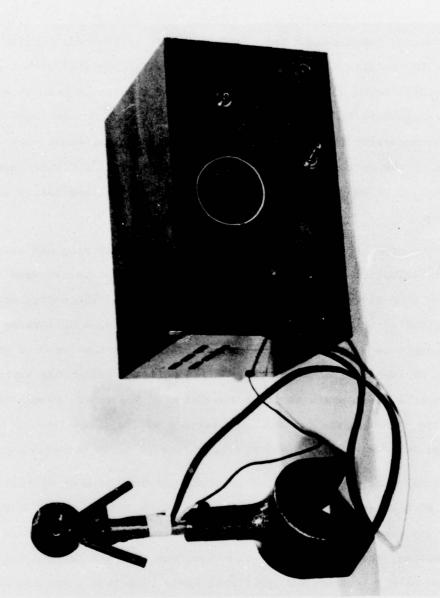
FIGURE 15 EFFECT OF TEMPERATURE RISE ON M-CENTER ABSORPTION PEAK
NaF SPECIMEN R-4

crystal by maintaining the specimen at or below the critical temperature of  $48\,^{\circ}\mathrm{C}$ .

Very little information is available on NaF but much has been written about the modification of the F center absorption bands in KCl. Both elevated temperatures and electric fields influence the color centers in the alkali halides. We have established that elevated temperatures below 48°C increase the M center absorption peak in NaF, but that above 48°C, the color centers become bleached. We therefore found it necessary to construct a specimen holder that would keep the crystal at the established critical temperature of 48°C during exposure to polarized uv and allow it to cool rapidly to room temperature after exposure.

The specimen holder consisted of a copper annular ring and surface plate to support the crystal. To heat the surface plate, we used a nichrome wire element supplied by a 12V, 1A source. The energy source was controlled by a variable voltage comparator using a calibrated thermocouple sensing element on the crystal surface that ensured precise control of the specimen temperature. The hollow annular ring surrounding the copper support plate could be flooded with ice water immediately after the specimen was exposed to polarized uv light and thereby lowering the specimen temperature to room temperature in a few seconds (Figure 16).

The NaF specimens used for this study had been colored by radiation with 1.96 MeV lithium ions to a fluence of 10.5 ion/cm<sup>2</sup>. The depth of coloration was approximately 4  $\mu$ m. Before exposure to temperature or uv radiation, the initial absorption spectrum for each crystal was recorded with a Beckman DK2 spectrophotometer. A test procedure was adopted whereby the crystal specimen was mounted in the holder, its temperature was raised to 48°C, after which it was exposed to polarized uv light at 364 nm for 1 to 2 minutes. At the power levels available, this exposure time represented 7 to 14 mJ or less than 5 percent of the



MA-1777-46

FIGURE 16 SPECIMEN HOLDER FOR TEMPERATURE CONTROL

power ordinarily required for writing or erasing in the specimen. The crystal was then immediately cooled to room temperature and the absorption spectrum recorded.

To compare the effects of exposure to temperature and uv radiation, we recorded the absorption curves for both vertically and horizontally polarized spectral scans. The specimen was then replaced in the temperature controlled mounting and maintained at 48°C for an additional 2 minutes. Both the M center and F center absorption peaks were affected by the exposure to temperature only (Figure 17). The increase in modulation between the vertical and horizontal scans was about 50 percent. Low levels of writing energy were used to make it possible to cycle the erase-write exposures without incurring permanent optical damage.

It has been suggested that the application of electric fields will also affect the M center absorption curves, particularly, if the specimen is maintained at an elevated temperature while the field is being applied. Because we did not have a suitable device for simultaneous application of both temperature and electric field, the two tests were made independently with the hope that a trend would become apparent. The electric field specimen holder was designed to support the specimen with copper electrodes along the diagonal of the square crystal. Voltage was applied to the electrodes from a variable voltage control box (Figure 18). An alternating current was applied to the crystal to avoid a directional electron drift and consequent removal of the color centers.

Studies with KCl indicate that if an electric field is applied to a crystal that has been additively colored to produce F centers, the crystal is highly insulating at room temperatures, but at elevated temperatures an electric current flow is observed in the crystal. When the current is applied, the colored portion moves away from the cathode

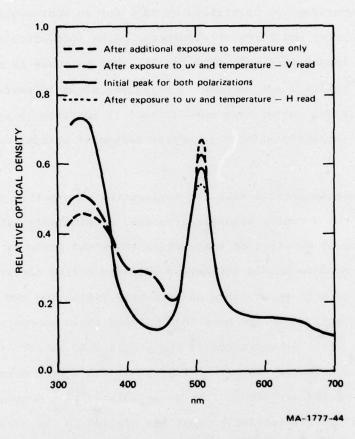


FIGURE 17 EFFECT OF TEMPERATURE ON M-CENTER ABSORPTION

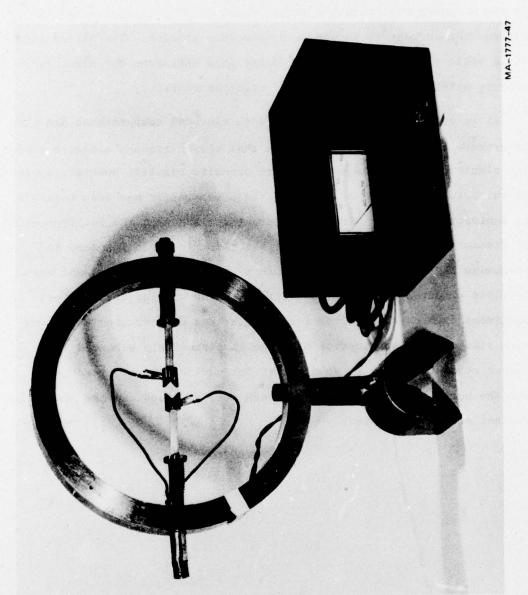


FIGURE 18 SPECIMEN HOLDER FOR ELECTRIC FIELD EXPERIMENT

and travels toward the anode, seeming to disappear into the electrode. The apparent mobility of the color centers under an applied electric field suggested that the F centers may be more inclined to alignment in the M center configuration during exposure to polarized uv if an alternating current field was simultaneously applied. The alternating field would not cause an F center drift into the anode but might well improve sensitivity by the enhanced electron mobility.

It is apparent from our studies with elevated temperatures and the literature on electric field effects that a simultaneous elevated temperature and electric field would produce more dramatic results; however, applying elevated temperature or an electric field separately has been valuable in indicating trends. Elevated temperatures enhanced the realignment of F centers to form M centers, but the electric field appeared to randomize the distribution of F centers, resulting in faster and more complete erasures with no marked effect on the writing cycle. Test specimens exposed to 9 mJ vertically polarized uv radiation at 364 nm with simultaneous application of an ac electric field enhanced the M center absorption peak to some extent for vertically polarized read, but the horizontally polarized read was not depressed and the modulation was not actually improved.

## III CONCLUSIONS

The ideal material for optical information storage would permit write, erase, and nondestructive readout in real time with archival retention of the recorded memory, if desired, with all functions being accomplished at moderate room temperatures and humidity. In addition, the material should be capable of recording at least 10 gray levels and a resolution limited only by the operating wavelengths and optical components. Sodium fluoride (NaF) has demonstrated all these properties with the single exception of sufficient sensitivity to approach real time response. Study of this material for holographic storage and recovery has shown that both bulk colored and ion implanted crystals can be used for information storage, each technique having distinctive characteristics that recommend specific application.

The initial effort of this study was aimed at using sodium fluoride crystals as an active element in an optical computer. However, the very poor sensitivity of this material precluded this application. Continuing studies suggested its use as the active memory element in an incoherent/coherent processor and possible applications for long term high density memories where writing speed was not a factor. Attempts to improve the writing sensitivity by temperature or electric field manipulation suggested a trend but did not predict sufficient improvement to achieve the initial goal for a real time device. In the continuing search for an efficient storage mechanism, the photodichroic alkali halides must be considered as possible candidates, because they possess most of the desirable properties for idealized optical storage.

#### IV REFERENCES

- 1. I. Schneider, M. Marrone, and M. Kabler, "Dichroic Absorption of M Centers as a Basis for Optical Information Storage," Applied Optics, Vol. 9, p. 1163 (May 1970).
- I. Schneider, M. Lehmann, and R. Bocker, "Extinction Technique for Optical Storage Using Anisotropic Color Centers in Alkali Halides," Applied Physics Letters, Vol. 25, No. 1, pp. 77-79 (July 1974).
- 3. H. Kogelnik, "Reconstruction Response and Efficiency of Hologram Gratings," Symposium on Modern Optics, Polytechnic Institute of Brooklyn (March 1967).
- 4. M. Lehmann, "Investigation of a Photodichroic Material for Holographic Storage and Recovery," Annual Technical Report, Office of Naval Research, Contract N00014-72-C-0260 (August 1974).
- 5. I. Schneider, "Information Storage Using Anisotropy of Color Centers in Alkali Halide Crystals," Applied Optics, Vol. 10, No. 4, pp. 980-981 (April 1971).
- 6. I. Schneider, private communication.
- 7. M. Lehmann, "Investigation of a Photodichroic Material for Holographic Storage and Recovery," Annual Technical Report, Office of Naval Research, Contract N00014-72-C-0260 (August 1973).
- 8. R. W. Ditchburn, "Light," (Interscience Publishers, London, 1963) pp. 569-570.
- I. Schneider, M. Lehmann, and R. Bocker, "Extinction Technique for Optical Storage Using Anisotropic Color Centers in Alkali Halides," Applied Physics Letters, Vol. 25, No. 1 pp. 77-79 (July 1974).
- Jenkins and White, "Fundamentals of Optics," (McGraw Hill Co., New York, 1957) p. 495.
- 11. M. Lehmann, "Investigation of a Photodichroic Material for Holographic Storage and Recovery," Annual Technical Report, Office of Naval Research, Contract N00014-72-C-0260 (August 1975).
- 12. I. Schneider, private discussion and demonstration at NRL (April 1975).
- 13. J. H. Schulman and W. D. Compton, International Series of Monographs on Solid State Physics. (Pergamon Press, London, 1962).